Final Report

Under the auspices of this funding, we have developed a program to synthesize and characterize highly monodispersed magnetic nanoparticles. We have been particularly interested in the origin of the exchange bias effect, which occurs in compound nanoparticles with a ferromagnetic core and an antiferromagnetic shell, and have mostly focused on Co/CoO core-shell nanoparticles. The exchange bias effect involves exchange coupling between the core moment and the antiferromagnetic shell which stabilizes the core moment, which would otherwise be quickly reorienting in ferromagnetic particles of this size.

Surprisingly, we found that the exchange bias effect is near the theoretical limit, as much as 100 times larger than that found in carefully engineered thin film systems. Further, the magnitude of the exchange bias effect does not depend on inter-particle spacing, proving that it is a local effect, satisfied within individual nanoparticles. We used TEM studies to prove that the core-shell interface is epitaxially smooth and highly oriented, indicating that the Co 100 surface is the most readily oxidized.

We have carried out neutron diffraction measurements on powders of Co/CoO nanoparticles. We observed broad nuclear peaks as well as four different magnetic peaks, from which we determined that the Neel temperature for the CoO shell is ~230 K, reduced from the bulk value for CoO of 293 K. We find that the magnetic structure consists of two different modulations. We find the conventional ½ ½ ½ modulation found in bulk CoO, which corresponds to Type II AF order with moments perpendicular to the 111 planes. Relative to an indicial nuclear peak, the ½ ½ ½ modulation has the same magnitude in three samples with different core and shell dimensions, and is identical to what is found in bulk CoO. We found a second magnetic modulation with a wave vector parallel to 100. This is almost forbidden in bulk CoO, but the matching tetragonal distortion in nanoparticle Co/CoO is more than 1000 times stronger. What is more, the 100 peak is much broader than the other peaks, indicating that it originates in only part of the CoO shell, most likely the interface itself. We proposed a simple structural model where the body centered Co in the bulk CoO structure is displaced, nominally by lattice mismatch and strain at the core-shell interface. The strain increases with oxidation, and we hypothesize that the moment induced at the interface improves the core-shell coupling and yields the enhanced exchange bias effect observed in these nanoparticle systems.

Intrigued by these results, we continued our neutron scattering investigations into inelastic measurements. Here we wished to study the evolution of the core dynamics as the system is cooled into the exchange biased state, which occurs near 200 K, well below the onset of antiferromagnetic order. We found that there was strong quasielastic scattering at high temperatures, which strengthens and even becomes resolution limited at the onset of antiferromagnetic order. Remarkably, we also observed a second and broader quasielastic peak which broadened below the Neel temperature. Both quasielastic peaks are replaced by inelastic scattering with the onset of the exchange bias freezing. The picture which emerges is that the inelastic peaks are spin waves , much as are found in bulk CoO. Neutron scattering experiments on a single crystal of CoO are ongoing to

test this hypothesis. The temperature dependence of the dynamics in the ordered state can be explained by a dynamically braking scenario, where the core slows down, and some of its kinetic energy is absorbed by the shell, which was initially at rest. We hypothesize that the onset of the exchange bias effect occurs which the core and shell are in the same dynamical frame.

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